Ultrasonic Characterizations of Slurries in a Bubble Column Reactor

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An indirect method (ultrasonic) and a direct technique were used to measure solid holdup in a bubble-column reactor. Nitrogen, water, and fine glass beads were used as the gas, liquid, and solid phases, respectively. The solid particle concentration in the slurry was varied from 5 to 30 wt %, and the gas superficial velocity was increased from 0.5 to 12 cm/s. The solid holdup measurements by the ultrasonic technique compared reasonably well with results obtained by the direct sampling technique.

Introduction

For the optimum design and operation of gas−liquid−solid three-phase reactors, the degree of dispersion of the solid (catalyst) in the reactor must be understood and controlled. The solid's distribution within the reactor greatly affects its performance. Because it is crucial to understand the influence of various reactions and reactor configurations on the solid holdup profile, measurement of solid holdup must be made under reaction conditions such as high temperature and pressure and with the presence of a reaction liquid medium. Various methods to measure this parameter, including optical, direct sampling, and static pressure methods, have already been developed and used experimentally. However, these methods have inherent disadvantages. Most of these optically based techniques use reflected, scattered, or absorbed light to measure the solid holdup. These methods are sensitive to the opacity of the slurry phase.[1] The direct sampling of solid particles from a slurry reactor suffers from either a need to perturb the reaction system or a difficulty of measurement because of high pressure and temperature.[2] The static pressure method also suffers some difficulties when the solid and liquid densities are close.[3] Accurate means of diagnosing the solid holdup, especially at industrially relevant conditions of high temperatures and pressures, are needed.

The ultrasonic technique offers more practical applications in that it is noninvasive, nondestructive, nonhazardous, rapid, and potentially applicable to high-temperature flow in high-pressure, opaque-wall reactors. However, the main disadvantage of this technique is that there are no commercially available ultrasonic instruments specifically designed to characterize solid holdup in a three-phase flow. Therefore, researchers have to develop their own instruments and data interpretation. The ultrasonic technique has been used widely in medical imaging, materials testing, flow detections, and level measurements.[4−6] The utilization of ultrasonic techniques for slurry characterizations has received considerable attention recently.[7−14] Some ultrasonic techniques are based on the principle of scattered acoustic pulses. The process is similar to sonar, in which bursts of acoustic energy are emitted into a liquid and reflections from discontinuities (solids and bubbles) are detected and measured. Abts and Dahi[7] have applied the ultrasonic technique to determine the concentration of oil droplets in an oil-recovery system by detecting the forward scattering of ultrasonic energy from oil droplets in the oil-recovery system. Foote[10] has reported that a particulate in a fluid can be identified by sending an ultrasonic pulse across the fluid, measuring the amplitude of the portion of the pulse scattered from a particulate at a preselected angle. The relative size of the particulate is determined from the magnitude of the scattered signal. An on-line ultrasonic particle monitoring for brewing operations has been reported by Behrman and Larson.[8] These ultrasonic techniques are based on the scattering of ultrasonic pulses. The use of the ultrasonic Doppler technique to characterize the local bubble rise velocity in a bubble column reactor has been investigated by Hilgert and Hofmann.[11] Bonnet and Tavlarides[9] have utilized an ultrasonic transmission technique to determine the dispersed-phase holdup of liquid−liquid dispersions by measuring the velocity of ultrasound in suspensions and emulsions. Tsouris et al.[13] have used an ultrasonic technique for real-time holdup monitoring to control extraction columns. Furthermore, Tsouris et al.[14] have applied a pulse-echo ultrasonic probe for local volume fraction measurements in liquid−liquid dispersions. Recently, a method involving the measurement of ultrasound transmission has been reported in a slurry-phase stirred-tank reactor which offers the possibility of using the ultrasonic technique to measure solid holdup in a three-phase slurry reactor.[15−17] The ultrasonic transmission uses measurements of the velocity and attenuation of the sound wave which travels directly through the slurry sample. When an acoustic wave strikes the boundary between two different media (liquid and solid) and the acoustic impedances of the two media are different, some acoustic energy will be reflected, some will be absorbed, and some will be transmitted. The reflected wave travels back through the incident medium (liquid) at the same velocity. The transmitted wave continues to move through the new medium (solid) at the sound velocity of the new medium. When the velocity of sound in a liquid (1496 m/s at 25 °C for water) is significantly different from that in a solid (5968 m/s at 25 °C for fused silica), a time shift (a velocity change) in the sound wave

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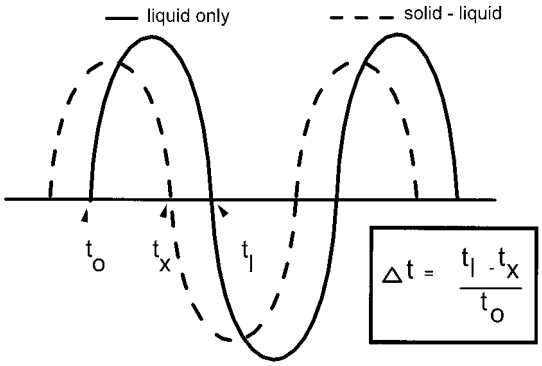

Figure 1. Effect of solids on the ultrasonic signal.

can be detected when solid particles are present relative to that for the pure liquid. The amplitude of the sound wave is also reduced when a solid particle is present because the wave is partially scattered and absorbed. Therefore, a change in amplitude of the sound wave can also be detected when solid particles are present relative to that for the pure liquid. Figure 1 shows how the detected sound wave is varied in time and in amplitude when solids are suspended in the liquid. The arbitrary first distinct zero crossing time in liquid and in solid−liquid are defined as t_l and t_x, respectively. The travel time between the transmitter and receiver in the liquid is defined as t_0. Okamura et al.[15] and Soong et al.[16,17] used a continuous stirred-tank reactor to correlate the solid holdup to the relative time shift $((t_l - t_x)/t_0)$. Thus, the solid holdup in a slurry reactor can be measured by sending an ultrasonic pulse across the slurry and measuring the amplitude and time shift of that portion of the transmitted pulse received at the opposite side of the reactor. Then, when the value is compared with those for a known concentration, the solid holdup is determined from the measured signal. The arbitrary first distinct zero crossing time, t_x, can also be determined in gas−liquid and gas−liquid−solid systems. Those researchers[15−17] also indicated that the t_x in the gas−liquid−solid system is not affected by the presence of gas bubbles and is fixed at the same position as that in the solid−liquid system. More recently, Soong et al.[18] has applied the ultrasonic technique to characterize slurries in a system consisting of molten paraffin wax, nitrogen, and glass beads at 189 °C. Furthermore, the application of the measurement of ultrasound transmission for gas holdup[19−21] and for gas holdup as well as the low concentration of solid (up to 1 wt %) under limited superficial gas velocities (up to 3 cm/s) in a slurry bubble-column reactor has been reported.[22,23] This leads to the initial study of using the ultrasonic technique for the measurement of solid holdup in a three-phase gas−liquid−solid bubble-column reactor over a wide range of superficial gas velocities and solid holdup and ultimately exploring this technique in a three-phase bubble-column reactor at elevated temperatures and pressures.

Experimental Method

A schematic representation of the bubble-column reactor in which the ultrasonic investigation was conducted is shown in Figure 2. The transparent acrylic bubble-column reactor has an internal diameter of 8.89 cm and a height of 290 cm. The column has six different axial locations for data collection. The ultrasonic signals are transmitted at 33 cm above the bottom of the gas distributor, which is a perforated-plate gas distributor with 15 × 1 mm diameter holes, along the center of the bubble-column reactor. Experiments were conducted in batch-mode operation (stationary liquid, water, and continuous flow of gas, nitrogen). Nitrogen bubbles were introduced through the gas distributor plate located at the bottom of the reactor. The nitrogen flow was controlled electronically to a maximum of 12 cm/s through a mass flow controller. Glass beads from Cataphote, Inc. (10−37 μm in diameter with a density of 2.46 g/cm^3), were used as the solid in the slurry. The solid holdup (solid weight/total slurry weight) was varied from 5 to 30 wt % for each nitrogen flow in the reactor. To evaluate the accuracy of the ultrasonic technique for solid holdup measurement, an independent slurry sampling device was installed. The measurement was conducted by inserting a stainless steel tubing (0.775 cm i.d.) horizontally into the center of the column at 0.635 cm above the path of the ultrasonic transmission. For each sampling, a 10 cm^3 of slurries sample was collected and analyzed for solid holdup characterization. The ultrasonic transmitter/receiver and the solid sampling device are positioned in such a way that both are measuring approximately the same hydrodynamic phenomena, as shown by the enlarged areas in Figure 2. The detailed information of the ultrasonic unit has been reported elsewhere.[16−18] Data were obtained with longitudinal waves at a frequency of 1 MHz using lithium niobate transducers (1.905 cm o.d.). Both the transmitter and receiver were mounted directly inside the reactor wall at 33 cm above the gas distributor.

Results and Discussion

Figure 3 illustrates the effects of the superficial gas velocity (SGV) on the transit time, t_x (an arbitrary first distinct zero crossing time in the ultrasonic signal; the details have been described elsewhere[17]), and on the gas holdup in the reactor. The average gas holdup was determined by visual observations of the expanded bed height versus the static bed height. During this process, we have visually identified the various flow regimes because our bubble column is transparent. Basically, three flow regimes were identified in the bubble column. The homogeneous or bubbly flow regime was observed when the SGV is 2.4 cm/s or less. The average gas holdup in this regime was found to increase linearly from 0.015 at a gas velocity of 0.26 cm/s to 0.093 at a SGV of 2.4 cm/s. The homogeneous flow regime is characterized by almost uniformly sized bubbles with equal radial distribution.[24] A transition flow regime exists between the SGV of 2.4 and 4 cm/s. The transition flow regime is characterized by large bubbles moving with high rise velocities in the presence of small bubbles. A slug flow regime is established when the velocity is 4 cm/s or higher. The average gas holdup increased from 0.1 to 0.148 when the flow regimes changed from transition to slug flow. The slug flow is characterized by large bubbles stabilized by the column wall, leading to the formation of bubble slugs for a column diameter of up to 15 cm. For the column diameter larger than 20 cm, a chun-turbulent flow regime will be established.[24] The transit time does not have an apparent correlation with the SGVs. It was approximately 72 μs at all SGVs and all flow regimes. The results suggest that the transit time of the ultrasonic signal is not affected by the nitrogen flow rate in the reactor within the current

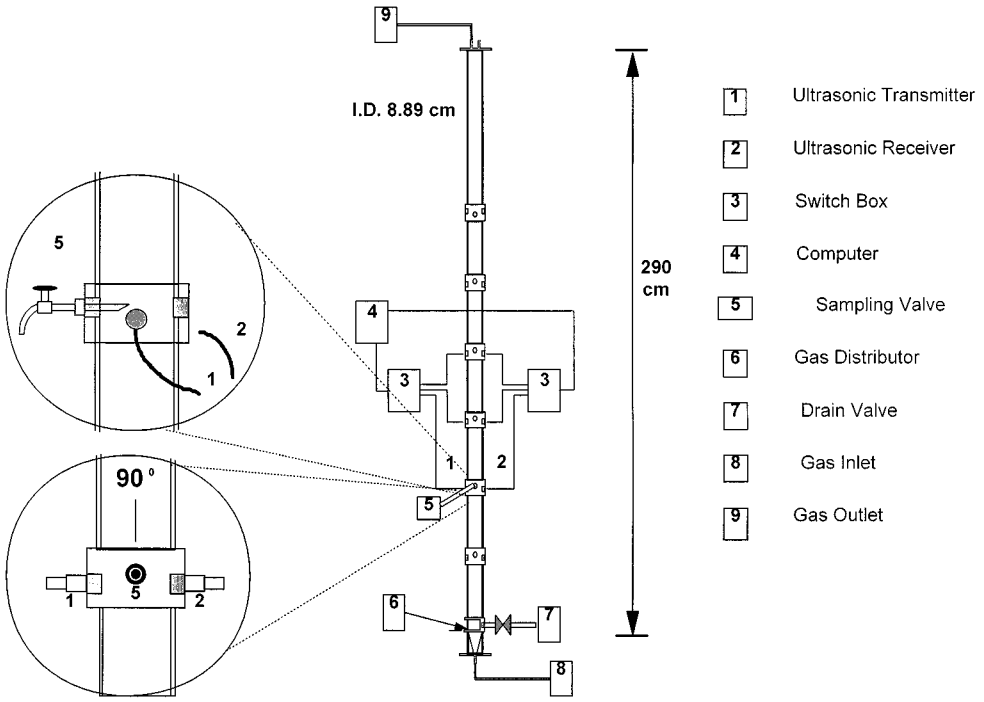

Figure 2. Schematic diagrams of the experimental setup.

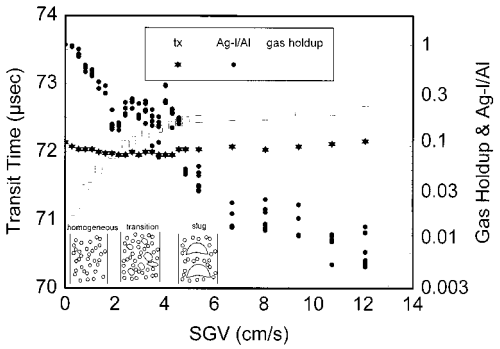

Figure 3. Effect of superficial gas velocity (SGV) on transit time, amplitude ratio (A_{g-l}/A_l), and gas holdup in the nitrogen/water system.

experimental conditions. What we measured was the signal not transmitted through the nitrogen. As discussed earlier, if the impedances of two media are widely separated, e.g., gas and liquid, then most of the energy is reflected back into the first medium (liquid) with little transmission into the second medium (gas). We measured the transmitted signals. Therefore, the transit time would not be affected by the presence of nitrogen flow. Chang et al.[19] also reported that the amplitude of the transmitted sound pulses depends significantly on the number of bubbles; however, the transit time does not change with the void fraction. Our results obtained from this study are similar to those of Chang et al.[19] for the air/water system. Uchida et al.[22] also measured the gas holdup in a bubble column for a gas−liquid system by determining the variation in transit time ratios. They indicate that the effect of gas holdup on the transit time is considerably small and is within the experimental errors. Therefore, measuring the variation in transit time ratios is not a precise way to determine the gas holdup. More recently, Warsito et al.[23] reported the measurement of gas holdup in a bubble column using the ultrasonic method. A change in transit time of 0.09 μs as the gas holdup increased from 0.05 to 0.1 was reported from their system. In a current study, we did not observe such a change in transit time as the gas holdup increased from 0.05 to 0.1 in our system. The discrepancy may be due to a different experimental setup or other factors. The small variation in transit time in this study is probably due to the experimental errors rather than the effect of nitrogen flow.

Figure 3 also shows the change in the amplitude ratio of the transmitted ultrasonic signals A_{g-l}/A_l and the local gas holdup in the reactor as a function of the SGV. A_{g-l} and A_l are the amplitudes of the transmitted signals with and without the presence of nitrogen, respectively. The transit time and amplitude measurements in this study are average values over five to seven different measurements. Each measurement consists of the time-averaged waveforms. A typical average of 100 waveforms is utilized in this study. Figure 3 suggests that the amplitude ratio is approximately an inverse exponential function of the gas holdup when the column is operated in the homogeneous flow regime (SGV of 2.4 cm/s or less). No discernible relationship between A_{g-l}/A_l and gas holdup could be found when the SGV is higher than 2.4 cm/s, i.e., while the column is operating in transition or slug flow regimes. As discussed earlier, the ultrasound beam cannot penetrate nitrogen bubbles that lie in its path; therefore, the amount of attenuation of the ultrasound beam by nitrogen bubbles is affected by the amount of gas volume fraction but also can be dependent on the bubble size present in the ultrasound path. When a large nitrogen bubble (slug) passes across the ultrasonic transmitted path, the transmitted signal will be reduced significantly. The transmitted signal will regain some amplitude immediately after the slug has passed through the transmitted path. Therefore, a large scatter of the A_{g-l}/A_l ratio is observed at SGV of 2.4 cm/s or higher.

The bubble sizes in the bubble column were also measured by the dual conductivity probe technique along with the ultrasonic measurements in separate

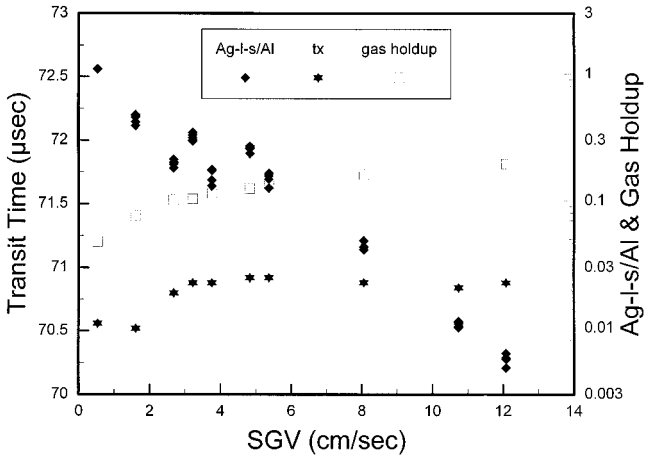

Figure 4. Effect of SGV on A_{g-l-s}/A_l, transit time, and gas holdup in the glass beads (30 wt %)/nitrogen/water system.

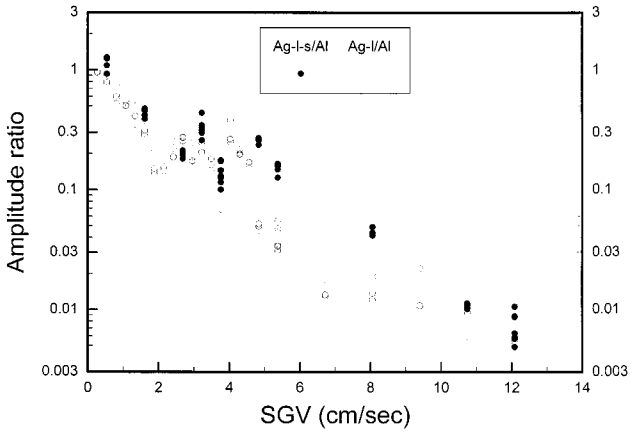

Figure 5. Comparison of the amplitude ratios, two-phase (A_{g-l}/A_l) versus three-phase (A_{g-l-s}/A_l) systems.

experiments.[21] The Sauter mean diameter is about 0.6 cm when the column is operated in the homogeneous flow regime. The Sauter mean diameter increases from 0.5 to 1 cm when the column is operated in the transition flow regime. Finally, the Sauter mean diameter is approximately 1 cm when the slug flow regime is established. The presence of nitrogen bubbles in the column may greatly affect its ultrasonic properties because of resonant scattering. The attenuation is greatest at frequencies near resonance.[6] The frequency at which resonance occurs depends on the physical properties of the component phases and the bubble size. For this study, the frequency utilized is not near the bubble resonance frequency. Therefore, the data collected in this study are not affected by the resonance effect.

Chang et al.[19] and Bensler et al.[20] measured void fractions in bubbly air−water two-phase flow using the ultrasonic transmission techniques. Chang et al.'s[19] results also showed that the A_{g-l}/A_l ratio has an exponential relationship with the void fraction and a function dependent on the bubble diameter. The effect of air bubble diameters on A_{g-l}/A_l ratios was found to be significant where A_{g-l}/A_l decreased with increasing bubble size. Bensler et al.'s[20] observations suggest that the A_{g-l}/A_l ratio has an exponential relationship with the interfacial area and the scattering cross section, which depends on the bubble radius (a) and the wavenumber (k) of the ultrasonic wave surrounding the bubble. Our observations of A_{g-l}/A_l in the nitrogen/water system are in qualitative agreement with those reported by Chang et al.[19] and Bensler et al.[20]

Figure 4 shows the effect of SGV on A_{g-l-s}/A_l, transit time, and average gas holdup in three-phase systems (30 wt % of glass bead/nitrogen/water) in the bubble-column reactor. A_{g-l-s} is the transmitted signal with the presence of solids and gas in liquid. A_l is the transmitted signal with the liquid only. Flow regimes' patterns similar to those in a two-phase system (Figure 3) were also observed in the three-phase system (Figure 4). The homogeneous flow regime was observed when the SGV is 2.68 cm/s or less. The average gas holdup in this regime was found to increase linearly from 0.047 at a gas velocity of 0.53 cm/s to 0.1 at a SGV of 2.68 cm/s. A transition flow regime exists between SGVs of 2.68 and 4 cm/s. A slug flow regime is established when the velocity is 4 cm/s or higher. The average gas holdup increased from 0.1 to 0.125 when the flow regimes changed from transition to slug flow. The fluctuation patterns of the amplitude ratio along with different flow regimes in Figure 4 are similar to those in Figure 3. For easy comparison, the amplitude ratios for two- and three-phase systems are presented together in Figure 5. In some instances, the values of the amplitude ratio are much higher and less scattered in the three-phase system, compared with those in the two-phase system. As mentioned earlier, the ultrasound beam *cannot* penetrate nitrogen bubbles that lie in its path; therefore, the amount of attenuation of the ultrasound beam by nitrogen bubbles is affected by the amount of gas in the form of bubbles present in the beam path for a two-phase system. When an ultrasonic pulse is sent across the slurry reactor, the amplitude of the pulse is *reduced* when it strikes a solid in the slurry because the pulse is partially scattered and absorbed. The scattering and viscous effects are often the predominant forms of attenuation in heterogeneous materials. The dominating mechanism depends on the range of ka where k is the ultrasound wavenumber and a is the radius of the particle. The scattering regime dominates when $ka \gg 1$ while the viscous regime governs for small particle size and lower frequencies ($ka < 1$).[25] For the present study, the ka ranges between 0.42 and 1.57. Thus, the dominating mechanism is between viscous and scattering effects. The latter occur when some of the ultrasonic wave is incident upon a discontinuity in a material; for example, a solid particle is scattered in directions which are different from that of the incident wave. The unscattered pulse is transmitted through the solid at the sound velocity of the solid and any nitrogen-bubble free path to the receiver. The reflected wave travels back through the water at the same velocity. The amplitude of the transmitted portion of the pulse is measured by the receiver located on the opposite side of the reactor. Therefore, the majority of the detected amplitude ratio in the slug flow regime for a three-phase system is larger than that from a two-phase system. Unlike the constant transit time observed in Figure 3 for a two-phase system, the transit time in a three-phase system may be related to the flow regimes operated and the presence of solids. The transit times were 70.56 and 70.52 μs respectively for superficial gas velocities of 0.537 and 1.611 cm/s when the column is operated in the homogeneous flow regime. In the transition flow regime, the transit time increased from 70.8 to 70.88 μs as the superficial gas velocity increased from 2.685 to 3.759 cm/s. The detected transit time is approxi-

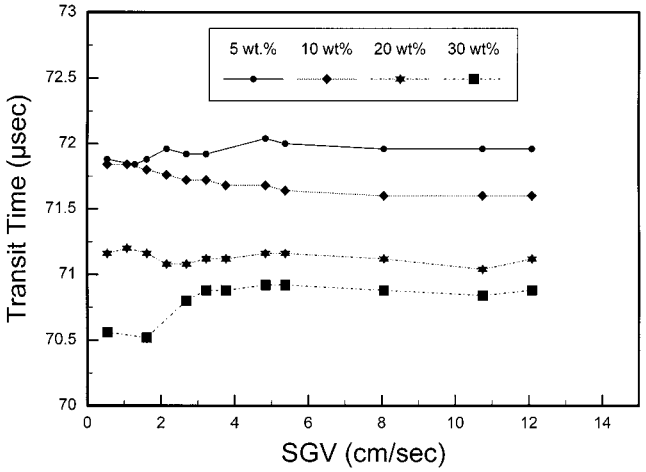

Figure 6. Effect of solids concentration on the transit time in the glass beads/nitrogen/water system.

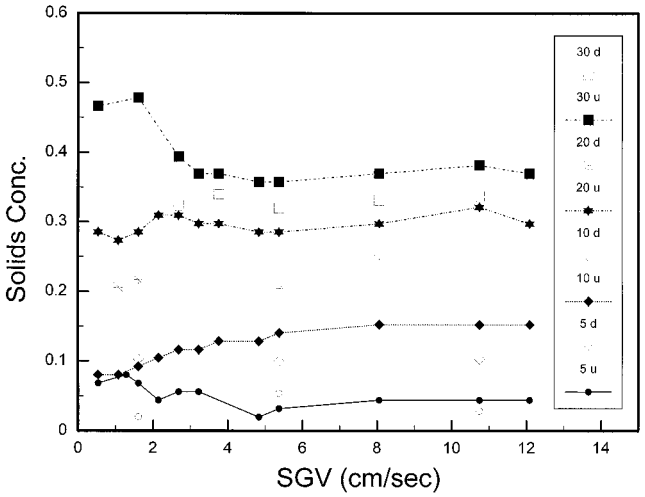

Figure 7. Solids concentration determined by ultrasonic and direct sampling techniques.

mately 70.88 μs for the slug flow regime, when the superficial gas velocity is 4 cm/s or higher. The fluctuation in transit time as flow condition changes can be understood by assuming that the presence of solid particles in the path of sound waves affects the transit time because sound waves travel at a much faster velocity in the solid than in water. The transit time of the transmitted ultrasonic pulse should depend on the quantity of solid particles present in the path. The higher the concentration of solid particles present in the path, the shorter the transit time that would be observed. The variation of transit time from 70.56 to 70.88 μs as the SGV changed from 0.537 to 12.05 cm/s suggests that there was variation in the concentration of solids in the ultrasonic path. For example, partial sedimentation occurred when the SGV was 2 cm/s or less. Thus, the concentration of solids should be high under these conditions. The detected constant transit time of approximately 70.88 μs in the slug flow regime suggests that there is a complete suspension of solids when the SGV is 4 cm/s or higher. Kölbel and Ralek[26] indicate that sedimentation will occur when the bubble column is operated under 2 cm/s or less and the complete suspension of the solid will be established when the column is operated at 4 cm/s or higher. Our ultrasonic observations are in good agreement with this finding.

Figure 6 illustrates the effects of solid holdup on the transit time measured at 33.65 cm above the gas distributor in the glass beads/nitrogen/water system at different SGVs. In this experiment, the SGV was systematically varied at any given initial solid holdup of 5, 10, 20, and 30 wt % in the bubble-column reactor. In general, the transit time varies with the variation of the superficial gas velocity of 4 cm/s or less at any given initial constant solid holdup loading in the reactor. The transit time was relatively constant when the SGV is 4 cm/s or higher. The transit times are around 71.96, 71.6, 71.12, and 70.88 μs for solid holdups of 5, 10, 20, and 30 wt %, respectively, when the SGV is 4 cm/s or higher. Therefore, the transit time can be utilized to determine the solid holdup when the column is operated in a complete suspension mode. The fluctuation of the transit time when the SGV is 4 cm/s or less may be attributed to both partial sedimentation and other factors which are under investigation.

The fractional change of transit time ($t/t_0 = (t_l - t_x)/t_0$) can be calculated on each individual transit time in Figure 6. From the fractional change of transit time, the solid holdup can be determined from the previously calibrated curve obtained from a stirred tank reactor (Figure 11 in Soong et al.[16]). The solid holdup determined from these procedures and the solid holdup determined by the direct sampling are illustrated in Figure 7. The solid holdup clearly varies with the SGVs. For a 30 wt % solid loading in the bubble column, the solid holdup varies from 0.466 to approximately 0.393 as the SGV increased from 0.53 to 2.68 cm/s. The occurrence of the partial semination in the column was also observed in the transparent acrylic column under this condition. At 2.685 cm/s gas velocity, the solid holdup determined by the ultrasonic technique is 0.393 compared to 0.323 by the direct sampling technique. At elevated SGV of 3.22 cm/s or higher, the solid holdup is approximately 0.369, as determined by the ultrasonic technique compared to 0.337 determined by the direct sampling technique. For 20 wt % solid loading in the bubble column, when the SGV increases to 3.22 cm/s or higher, the ultrasonic technique estimates the solid holdup at 0.295. However, the solid holdup determined by the direct sampling technique is between 0.2044 and 0.246 under these conditions. For 10 wt % solid loading in the bubble column, the holdup profile is similar to that of 20 wt %. At the SGV of 1.61 cm/s, the solid holdup determined by the ultrasonic technique is 0.092 compared with that of 0.102 determined by direct sampling. For 5 wt % loading conditions in the bubble column, when the SGV is 3.22 cm/s or higher, the solid holdup is approximately 0.044, as determined by the ultrasonic technique. Furthermore, the solid holdup determined by direct sampling is between 0.0539 and 0.0286 under the same experimental conditions. The solid holdup measurements by the ultrasonic technique compared reasonably well with results obtained by the direct sampling techniques. Some discrepancies observed between these two techniques are probably due to the nature of these techniques. The ultrasonic technique measures the average solid holdup in the ultrasound path while direct sampling determines the collected local solid holdup. The simple time-averaged method utilized in this study does not account for the sound refraction and reflection of the solid phase on the transit time. As suggested by Tavlarides et al.,[9,27,28] those effects may affect the measured travel time.

Wang and Chang[29] have reported the monotonic behavior on the acoustic attenuation in a solid−water two-phase flow system. The critical mass fraction is dependent upon the particle size and frequency and is around 20−45 wt %. Urick[30] conducted the acoustic study in the silica particles in a water system. Urick[30] has proposed a phenomenological approach to correlate the solids concentration and velocity of sound through the medium. He predicts a minimum in sound speed (which is related to the reverse of the transit time) at a solid fraction of about 25−30 wt %, and the prediction is based on a ka value of 3.4×10^{-4}. Results obtained from the current study (Figures 6 and 7) show different behavior. The transit time decreases monotonically with solids concentration. Our current nonmonotonic behavior as a function of solids concentration is probably due to the ka (between 0.42 and 1.57) spanning 3−4 orders of magnitude and other factors. Uchida et al.[22] and Warsito et al.[23] have reported some experimental results obtained from low concentrations of solids (up to 3 wt %) and low gas velocities (up to 3 cm/s). It is difficult to compare our data collected under high gas velocities and high solid holdup with those collected under different conditions. Warsito et al.[23] also proposed a theoretical model that related the solid holdup with the transit time ratio. The coupled-phase model of suspension approaches have also been investigated by Harker and Temple.[31] These approaches can be applied to further theoretical study.

Conclusions

An ultrasonic transmission technique has been developed to measure solid holdup in a gas−liquid−solid bubble-column reactor. The results presented in this study show that the transit time of an ultrasonic signal is influenced by the variation of solid holdup and the operating conditions in the bubble column. The transit time can be correlated to the solid holdup. The variation of nitrogen flow has little influence on the observed transit time within the two-phase flow conditions studied. The ultrasonic technique is potentially applicable to high-temperature, nontransparent fluids in high-pressure, metallic reactors and, with some modifications for solid holdup measurements, is applicable in slurry bubble-column reactors.

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Disclaimer

References in this report to any specific commercial product, process, or service is to facilitate understanding and does not necessarily imply its endorsement or favoring by the United States Department of Energy.

Nomenclature

a = bubble radius or particle radius, m
A_l = amplitude of the transmitted signal in the liquid
A_{g-l} = amplitude of the transmitted signal in the gas−liquid system
A_{g-l-s} = amplitude of the transmitted signal in the gas−liquid−solid system
k = wavenumber of the ultrasonic wave which surrounds the bubble
t_l = arbitrary first distinct zero crossing time in liquid, s
t_x = arbitrary first distinct zero crossing time in a gas−liquid, solid−liquid, or gas−liquid−solid system, s
t_0 = arbitrary travel time of the sound wave between the transmitter and the receiver in liquid, s

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